



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/US99/29986 <b>(22) International Filing Date:</b> 17 December 1999 (17.12.99) <b>(30) Priority Data:</b> 60/113,132 21 December 1998 (21.12.98) US <b>(63) Related by Continuation (CON) or Continuation-in-Part (CIP) to Earlier Application</b> US 60/113,132 (CON) Filed on 21 December 1998 (21.12.98) <b>(71) Applicant (for all designated States except US):</b> MAGNA INTERNATIONAL OF AMERICA, INC. [US/US]; 600 Wilshire Drive, Troy, MI 48084 (US). <b>(72) Inventor; and</b> <b>(75) Inventor/Applicant (for US only):</b> WILSON, Phillip, S. [US/US]; 5480 Huron Hills Drive, Commerce Township, MI 48382 (US). <b>(74) Agents:</b> BARUFKA, Jack, S. et al.; Pillsbury Madison & Sutro LLP, 1100 New York Avenue, N.W., Washington, DC 20005 (US).		<b>(81) Designated States:</b> AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
<b>(54) Title:</b> METHOD OF MAKING ROTATIONALLY MOULDED PARTS HAVING NANO-PARTICLE REINFORCEMENT <b>(57) Abstract</b> <p>A method of producing reinforced articles comprises (a) preparing a powder composition of at least one thermoplastic and about 2 % to about 15 % by volume reinforcing particles. The reinforcing particles are substantially uniformly dispersed in the thermoplastic. At least 50 % of the reinforcing particles are less than about 20 layers thick, the layers of the reinforcing particles having a unit thickness of between about 0.7 nm – 1.2 nm; (b) heating the powder into a molten material; (c) rotating the molten material in a mold cavity so that the molten material conforms to the surfaces of the mold cavity, the molten material having substantially the same uniformity of dispersion of the reinforcing particles therein in comparison with the uniformity of dispersion of the reinforcing particles in the thermoplastic powder; and (d) cooling the molten material to form a reinforced article.</p>		

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## METHOD OF MAKING ROTATIONALLY MOULDED PARTS HAVING NANO-PARTICLE REINFORCEMENT

5      BACKGROUND OF THE INVENTION

Rotational molding of polymers offers an economical approach to producing hollow articles for various applications. Traditionally, injection molding has been used to produce complex, higher cost molded items at high production rates, whereas blow molding is employed for items such as one-piece closed vessels. Rotational molding is ideally suited for producing hollow, closed, relatively thick-walled items at reasonably low per unit cost. Advantages of rotational molding include uniform wall thickness of the molded articles, as well as close control of the weight of molded products.

10      During rotational molding, thermoplastic resin powder of relatively low viscosity is placed inside an aluminum mold and the mold is heated in an oven. The mold is rotated during heating, thereby forcing the resin against the mold surface. The resin then sinters and fuses, coating the inside of the mold. The melt is then cooled and solidifies to assume the shape of the mold. Articles of uniform wall thickness, zero orientation and good physical properties may be generated via this process.

15      Certain automobile parts may be efficiently produced by rotational molding methods. Such parts historically have included air ducts, fuel tanks, glove boxes and fender liners. Solid removable convertible tops, such as removable hard tops for sport utility vehicles, may also be rotationally molded. In an attempt to reduce automobile unit weight, the automotive industry has sought techniques with which to reduce wall thickness of molded automobile parts without compromising impact resistance and dimensional strength. Additionally, greater wall thickness of such molded articles requires more raw materials per molded part, thereby increasing the cost of production. Cycle times may be high for rotational molding due to the heating and cooling periods required, which depend upon the wall thickness of the article being formed.

25      Conventional reinforced plastics have not been used effectively in rotational molding due to non-uniform dispersion of the reinforcement fibers. Large

rotationally molded articles, articles with thin walls, and rotationally molded articles requiring good impact strength tend to require thermoplastic resin compositions having reinforcing materials incorporated therein. Glass reinforcement fibers have been added to molds containing thermoplastic resin powders for reinforcement purposes. During the rotational molding process, melting of thermoplastic powder, combined with centrifugal force tends to somewhat separate the thermoplastic or resin from the glass fibers, as the resin powder migrates towards the mold surfaces, while the glass fibers tend remain toward the center of the die cavity. Thus, uniform distribution of the glass fibers throughout the molded article is difficult to achieve. The surface of the mold becomes highly resin rich with few glass fibers present. The resulting molded articles lack strong reinforcement because the fibers do not fully intermix with the resin in an even fashion.

U.S. Patent number 4,363,687 to Anderson discloses a method of producing large, fiber reinforced storage tanks. The reinforced resin articles of Anderson '687 contain glass reinforcement fiber, resin, and catalyst which are applied to the inner surface of a revolving mold. As described hereinabove, this traditional approach to reinforced rotational molding causes resin rich regions to form on the surface of the mold due to uneven dispersion of the glass fibers during rotation.

Filament winding around or within molded articles are additional techniques used to reinforced rotationally molded products. For example, U.S. patent number 4,123,307 to Lemelson is directed to a method of reinforcing a molded article whereby filamentary material is wound around the article, thus strengthening the outer stratum thereof. U.S. patent number 4,002,715 to Usui et al. discloses a synthetic resin pipe which is reinforced by winding belt-like or thread-like glass fiber around a tapered core bar, inserting the arrangement into a tapered mold, and thereafter removing the core bar such that the wound glass fiber remains. Thermosetting liquid resin is then introduced into the mold, which is rotated at an inclined angle. Rotation of the mold causes the resin material to permeate the reinforcing material, which then hardens into the form of a pipe. The cited techniques using external and preformed fibers do not permit even reinforcement throughout the entire molded article.

Therefore, a need exists for improved reinforcement of hollow rotationally molded resin articles.

### SUMMARY OF THE INVENTION

5           An object of the present invention is to overcome the problems delineated hereinabove. In accordance with this object, the present invention provides a rotationally molded article. Such articles are suitable for use as automobile components. The articles are formed by a method comprising (a) preparing a powder composition of at least one thermoplastic and about 2% to about 15% by  
10           volume reinforcing particles. The reinforcing particles are substantially uniformly dispersed in the thermoplastic. At least 50% of the reinforcing particles are less than about 20 layers thick, and at least 99% of the reinforcing particles are less than about 30 layers thick, the layers of the reinforcement particles have a unit thickness of between about 0.7 nm - 1.2 nm; (b) heating the powder into a molten material;  
15           (c) rotating the molten material in a mold cavity so that the molten material conforms to the surfaces of the mold cavity, the molten material having substantially the same uniformity of dispersion of the reinforcing particles therein in comparison with the uniformity of dispersion of the reinforcing particles in the thermoplastic powder; and (d) cooling the molten material to form a reinforced  
20           article.

          The parts manufactured according to the invention are constructed to be both lightweight and strong, exhibiting good impact resistance and dimensional stability.

          This and other objects of the invention can be more fully appreciated from  
25           the following detailed description of the preferred embodiments.

### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

          In accordance with the present invention, reinforcing nanoparticle fillers are added in levels of only a few percent by volume to polymer resin compositions  
30           prior to rotational molding into an article. As a result, the impact resistance and dimensional stability of thin-walled molded articles made of resins, such as polyolefins, is improved. For example, large, hollow rotationally molded articles

can be reinforced according to the invention, thereby imparting good dimensional stability.

FIG. 1 illustrates a conventional rotational molding apparatus, generally indicated at 10, that can be employed in the method of the present invention. The apparatus 10 includes a mold structure 12 defining a mold cavity 14. The mold structure 12 is mounted for rotation around an axis extending through a feeding tube 16. More specifically, the opposite ends of the mold structure 12 are mounted on bearing carrying supports 18. One end of the mold structure is connected by linkage 20 that connects the structure 12 to a motor 22 that drives the mold structure 12 for rotation about said axis. The motor 22 and supports 18 are mounted on a mounting structure 24. The mounting structure 24 is mounted on a movable mechanism 26 that can tilt the mounting structure 24 to ensure that powder introduced into the cavity 14 is uniformly distributed along the longitudinal axis.

The aforementioned tube 16 is hollow and provided with a plurality of openings 30. A powder material, to be described in greater detail below, is introduced through a feeding tube 32 into the interior of tube 16. The powder can then be distributed into the cavity 14 through the openings 30. In an alternate configuration, the present invention contemplates that the mold structure 12 can be provided with two mold halves (or parts) or be provided with a door that can be opened in order to deposit a pre-measured amount of powder into the cavity 14. In this alternate construction, tube 16 would not be provided.

After the powder material is introduced into the cavity 14, the die structure 12 is rotated by motor 22 about the axis. The die structure is provided with a spiral passageway, as indicated at 44, or otherwise jacketed to enable hot oil or other hot liquid to be used to heat the die structure. Alternately, the passageway 44 can be omitted, and the entire structure 12 placed in an oven.

The powder is heated so that it is in molten form. The molten material is then evenly dispersed over the interior surface 46 of the mold structure 12. After an appropriate amount of time, cooling fluid, such as water, may be introduced into the passageway to cool the molten material to solidify the material as it continues to rotate. The solidified article is then removed from the structure 12.

In accordance with the present invention, and as will be described in greater detail below, the powder used preferably comprises at least one thermoplastic and about 2% to about 15% by volume reinforcing particles. The said reinforcing particles (or "nanoparticles" as described below) are substantially uniformly dispersed in the thermoplastic, at least 50% of the reinforcing particles being less than about 20 layers thick, the layers of the reinforcing particles having a thickness of between about 0.7 nm - 1.2 nm.

Preferably, the powder introduced into the mold cavity is formed by grinding of pellets, the pellets preferably formed in an extruding process. More specifically, the nanoparticles and the thermoplastic material are preferably extruded together to form the pellets. The pellets are then ground in a mechanical grinder to form the powder. The nanoparticles are substantially uniformly dispersed in the pellets and hence also in the particles forming the powder. Because the nanoparticles are very small, there is no possibility of such reinforcing particles becoming damaged in the grinding operation. This is unlike a methodology that would use larger reinforcements, such as glass fibers, which reinforcements would have their reinforcing characteristics damaged by a grinding operation.

In an alternate embodiment, the thermoplastic is originally formed as a powder and the nanoparticles are later added to the powder in a powder blending device.

The parts manufactured in accordance with the present invention comprise a composite material of a polymer having dispersed therein reinforcement fillers in the form of very small mineral reinforcement particles. The reinforcement filler particles, also referred to as "nanoparticles" due to the magnitude of their dimensions, each comprise one or more essentially flat platelets. Generally, each platelet has a thickness of between about 0.7-1.2 nanometers. The average platelet thickness is approximately 1 nanometer.

The preferred aspect ratio of the reinforcement particles, which is the largest dimension of a particle divided by the thickness of the particle, is about 50 to about 300. At least 80% of the particles should be within this range. If too many particles have an aspect ratio above 300, the material becomes undesirably viscous for forming parts in an effective and efficient manner. If too many particles have an

aspect ratio of smaller than 50, the particle reinforcements will not provide the desired reinforcement characteristics. More preferably, the aspect ratio for each particle is between 100-200. Most preferably at least 90% of the particles have an aspect ratio within the 100-200 range.

5           The platelet particles or nanoparticles are derivable from larger layered mineral particles. Any layered mineral capable of being intercalated may be employed in the present invention. Layered silicate minerals are preferred. The layered silicate minerals that may be employed include natural and artificial minerals. Non-limiting examples of more preferred minerals include  
10   montmorillonite, vermiculite, hectorite, saponite, hydrotalcites, kanemite, sodium octosilicate, magadite, and kenyaite. Mixed Mg and Al hydroxides may also be used. Various other clays can be used, such as claytone H.Y. Among the most preferred minerals is montmorillonite.

          To exfoliate the larger mineral particles into their constituent layers,  
15   different methods may be employed. For example, swellable layered minerals, such as montmorillonite and saponite are known to intercalate water to expand the inter layer distance of the layered mineral, thereby facilitating exfoliation and dispersion of the layers uniformly in water. Dispersion of layers in water is aided by mixing with high shear. The mineral particles may also be exfoliated by a shearing process  
20   in which the mineral particles are impregnated with water, then frozen, and then dried. The freeze dried particles are then mixed into molten polymeric material and subjected to a high sheer mixing operation so as to peel individual platelets from multi-platelet particles and thereby reduce the particle sizes to the desired range.

          The polymer composites of the present invention are prepared by combining  
25   the platelet mineral with the desired polymer in the desired ratios. The components can be blended by general techniques known to those skilled in the art. For example, the components can be blended and then melted in mixers. Thereafter, solidified polymer/nanoparticle mixtures may be ground into powders for rotational molding. The extremely small size of the nanoparticles permits such grinding  
30   without loss of reinforcing capabilities. The ratios will be determined based on, for example, desired dimensional stabilization and/or desired mechanical properties of the final molded article.



Additional specific preferred methods, for the purposes of the present invention, for forming a polymer composite having dispersed therein exfoliated layered particles are disclosed in U.S. Patent Nos. 5,717,000, 5,747,560, 5,698,624, and WO 93/11190, each of which is hereby incorporated by reference. For  
5 additional background, the following are also incorporated by reference: U.S. Patent Nos. 4,739,007 and 5,652,284.

Typical resins used in rotational molding include vinyl plastisols and powdered low viscosity polyolefins. Liquid vinyl dispersions may be used to rotationally cast automotive parts, such as arm rests. Other automobile components  
10 may be molded of polyolefins. Powdered polyolefins are available in a variety of melt indexes, densities and particle sizes. For example, polyethylenes having low melt indexes provide for articles having good impact strength as well as resistance to low-temperature brittleness. High density resins impart high rigidity to rotationally molded products, permitting reduction of wall thicknesses of the  
15 products. Increasing the density of the polyolefin resin also raises the melting point of the polymer, permitting higher service temperatures to be used.

Preferably, the thermoplastic of the present invention is a powdered polyolefin or a homogenous or copolymer blend of polyolefins. The preferred polyolefin is at least one member selected from the group consisting of  
20 polypropylene, ethylene-propylene copolymers, thermoplastic olefins (TPOs), and thermoplastic polyolefin elastomers (TPEs). The process permits use of recycled materials, such as scrap polyolefins or post consumer polyolefins. For high performance applications, engineering thermoplastics are most preferred. Such engineering thermoplastic resins may include polycarbonate (PC), acrylonitrile  
25 butadiene styrene (ABS), a PC/ABS blend, polyethylene terephthalates (PET), polybutylene terephthalates (PBT), polyphenylene oxide (PPO), or the like.

The exfoliation of layered mineral particles into constituent layers need not be complete in order to achieve the objects of the present invention. The present invention contemplates that at least 50% of the particles should be less than about  
30 20 nanometers in thickness. Otherwise stated, more than about 50% of the particles should be less than about 20 platelets (20 layers) stacked upon one another in the thickness direction. In addition, at least 99% of the reinforcement particles should be less than about 30 layers (i.e., 30 nanometers) in thickness. Preferably, at least

90 % of the particles should have a thickness of less than 5 layers. It is most preferable to have as many particles as possible to be as small as possible, ideally including only a single platelet. Particles having more than 30 layers may behave as stress concentrators and are preferably avoided.

5           Generally, in accordance with the present invention, each of the automotive parts that can be manufactured in accordance with the principles of the present invention should contain nanoparticle reinforcement in amounts less than 15% by volume of the total volume of the part. The balance of the part is to comprise an appropriate thermoplastic material and optionally, suitable additives. If greater than  
10   15% by volume of reinforcement filler is used, the viscosity of the composition becomes too high and thus difficult to mold. Preferably, the amount of reinforcing nanoparticles is greater than 2% by volume (as lower amounts would not achieve the desired increase in strength) and less than 15%.

          Preferably, rotationally molded automobile components comprise  
15   reinforcement particles of the type described herein at about 2-15% of the total volume of the part, with the balance comprising the thermoplastic substrate. It is even more preferable for these reinforced components to have reinforcement particles of the type contemplated herein comprising about 3%-8% of the total volume of the part. For some applications, inclusion of about 3%-5% reinforcing  
20   nanoparticles is optimal.

          When about 90% of the nanoparticles in the composition are less than 5 nm in thickness, a more preferred uniform distribution of the particles occurs in the resin, which translates into evenly distributed reinforcement particles throughout the mold cavity. The extremely small size of these reinforcing particles permits  
25   them to uniformly disperse with powdered resin when subjected to centrifugal forces in a rotating mold. A reduction to near elimination of unreinforced surface regions occurs in the final molded product, accordingly.

          In addition to reinforcing agents, other additives may optionally be included in the polymer resin composition to improve processability. For example, aging  
30   modifiers, such as glycerol monostearate, are useful additives in polymer compositions for molding. Aging modifiers are typically present in an amount from about 0.5% to about 5% polyolefin resin. Other additives include pigments, heat stabilizers, antioxidants, flame retardants, ultraviolet absorbing agents and the like.

Reinforced articles of the invention exhibit improved properties over non-reinforced articles. For example, polyethylene articles having 5% nanoparticles by volume, wherein 90% of the particles have 5 or fewer layers, increased flexural modulus by 2.5 to about 3 times over compositions lacking reinforcing nanoparticles, as measured under ASTM D790 test conditions. This 5% nanoparticle polyethylene article exhibited > 200% elongation to rupture. By contrast, about 25% glass fiber reinforcement is required in such articles to achieve an equivalent modulus. Polypropylene articles according to the invention showed about a 60% improvement in flexural modulus over articles lacking reinforcement nanoparticles. Thus, the use of reinforcing nanoparticles according to the invention provides articles having good flexural stiffness.

In the method of the present invention, producing reinforced articles comprises preparing a powder composition of at least one thermoplastic powder and about 2% to about 15% by volume reinforcing particles. The reinforcing particles are substantially uniformly dispersed in the thermoplastic powder. At least 50% of the reinforcing particles are less than about 20 layers thick, with the layers comprising platelets which have a thickness of between about 0.7 nm to 1.2nm.

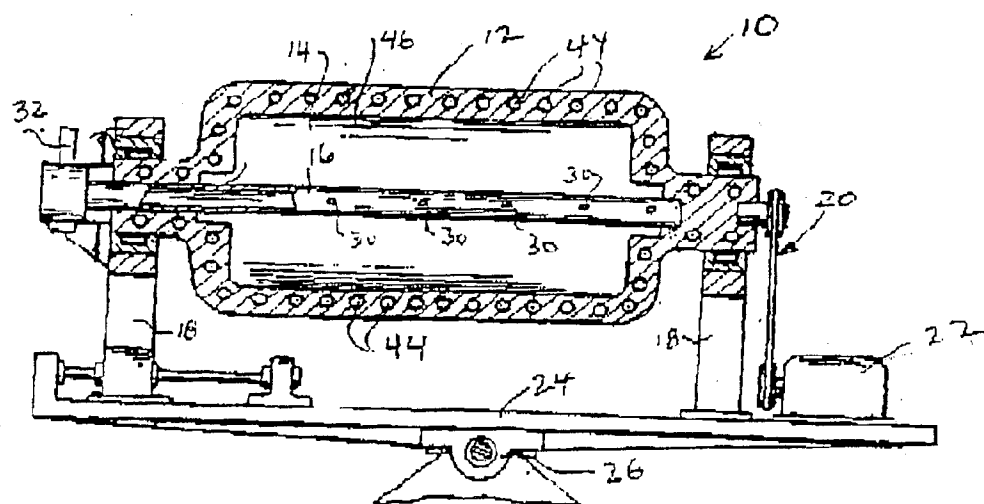
The method of producing reinforced articles further comprises heating the powder into a molten material, rotating the powder in a mold cavity so that the molten material conforms to surfaces of the mold cavity, and cooling the molten material to form a reinforced article. The molten material has substantially the same uniformity of dispersion of the reinforcing particles therein in comparison with the uniformity of dispersion of the reinforcing particles in the thermoplastic powder.

The relatively small size of the nanoparticles enables the reinforcement particles to be dispersed uniformly in the molded article, irrespective of the centrifugal force applied in rotational molding. The uniformity of dispersion in the molten powder is essentially the same as the uniformity of dispersion of the nanoparticles in the thermoplastic powder.

It should be appreciated that the foregoing description is illustrative in nature and that the present invention includes modifications, changes, and equivalents thereof, without departure from the scope of the invention.

What is claimed is:

1. A method of producing reinforced articles comprising:
  - (a) preparing a powder composition of at least one thermoplastic and about 2% to about 15% by volume reinforcing particles, said reinforcing particles being substantially uniformly dispersed in the thermoplastic, at least 50% of the reinforcing particles being less than about 20 layers thick, and at least 99% of the reinforcing particles being less than about 30 layers thick, the layers of said reinforcing particles having a unit thickness of between about 0.7 nm - 1.2 nm;
  - (b) heating the powder into a molten material;
  - (c) rotating the molten material in a mold cavity so that the molten material conforms to the surfaces defining the mold cavity, said molten material having substantially the same uniformity of dispersion of said reinforcing particles therein in comparison with the uniformity of dispersion of said reinforcing particles in said thermoplastic powder; and
  - (d) cooling the molten material to form the reinforced article.
2. A method according to claim 1, wherein said preparing of said powder composition is accomplished by grinding of pellets, said pellets comprising said at least one thermoplastic and said about 2% to about 15% by volume reinforcing particles, said reinforcing particles being substantially uniformly dispersed in the pellets.
3. A method according to claim 2, wherein said pellets are formed by extrusion.

FIG. 1

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 99/29986

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 B29C67/00 B29C41/04 C08K3/34

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 B29C C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4 739 007 A (OKADA AKANE ET AL) 19 April 1988 (1988-04-19) column 2, line 50 -column 3, line 65 ----	1-3
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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## INTERNATIONAL SEARCH REPORT

Int'l Application No  
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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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information on patent family members

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